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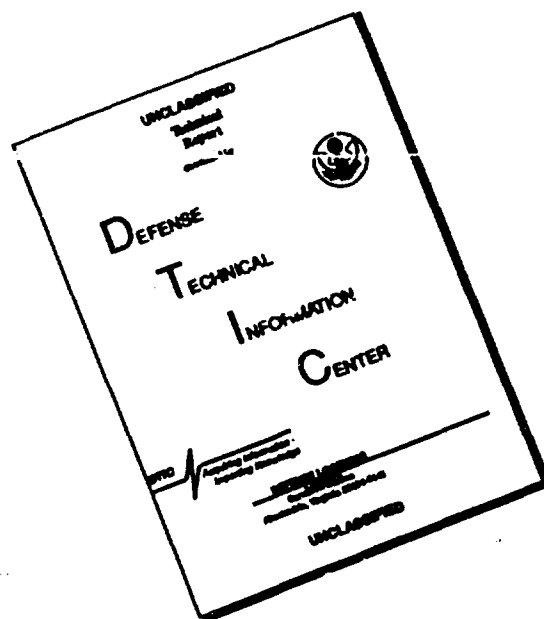
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AEROSOLS AS CARRIER OF NATURAL RADIOACTIVITY

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ABSTRACT

Description of an apparatus for ionizing natural aerosol and for classifying the particles by mobility. The natural β -activity of the particles is utilized as indicator. It was shown that particles with a radius between 1 and 15×10^{-7} cm carry 65% of the activity. No indications were found that a particular size classification of particles is preferred. Comparisons are made with similar measurements by Wilkening in New Mexico.

Introduction

In connection with a number of studies carried out in our Institute on natural and artificial activity of the atmosphere (Ref. 1), it appeared desirable to investigate the distribution of activity over the size range of the aerosol. Such measurements have already been made by Wilkening in New Mexico for natural atmospheric activity which is about 5 times higher in New Mexico than in Munich. At the start of our measurements in 1959, the artificial atmospheric activity due to nuclear-weapons tests was very low so that we initially investigated only the natural activity. This amounted during the period of measurement to approximately 100 to 1,000 times of the artificial activity, i.e. in the mean 1 to 2×10^{-10} Curie of radium B + C and 2 to 4×10^{-12} Curie of thorium B + C.

It was of particular interest whether the distribution of activity, on which Wilkening concluded from his measurements, over only two discrete classes of particle size, i.e. diameters of 0.9 and/or 1.8×10^{-6} cm, existed also in Munich. It may be assumed that the various conditions in New Mexico and/or Munich responsible for the origin and size distribution of the aerosol had an influence also on the apposition of the radium and thorium metabolons. We therefore designed an apparatus essentially corresponding to that of Wilkening and suitable for investigating approximately the same range of mobility.

Measuring Arrangement

The apparatus (Fig. 1) consists of a cylindrical condenser which the air enters by a small ring gap along the inner electrode. The latter consists of brass with a diameter of 1.3 cm and is at ground potential. The outer electrode with an inner diameter of 4 cm consists of 10 nickel-coated brass rings which can be assembled into a tube with a length of 40 cm. The electrode potential can be varied between + 0.6 and + 3 kV.

Two laminar air streams are passed through the cylindrical condenser. Room air from which activity has been removed by a cotton filter, flows along the outer electrode and serves to keep the active outer-air stream, entering parallel to the inner electrode, laminar. Outer air is drawn in by a grounded brass tube with an internal diameter of 1.5 cm. It flows around a tungsten coil serving as ionizer which is charged to 6 kV in relation to the ground. In the vicinity of the tungsten coil, the aerosol particles contained in the air are in large part charged negatively by passing through the negative space charge of the corona discharge. The air stream then enters the cylindrical condenser through a ring gap, 6 cm long and 1 mm wide. The laminar character of the double stream is observed with the aid of cigarette smoke through a glass tube placed in the location of the anode. We determined experimentally the most favorable values for the double stream, i.e. optimum large Q_2 , as follows: non-active air $Q_1 = 6.2 \text{ m}^3/\text{hr}$; active outer air $Q_2 = 0.6 \text{ m}^3/\text{hr}$. The volume of flow was measured with a gas-flow meter.

the aerosol particles leave the ring gap, they move in the anode under the action of the electric field. According to Becker (Ref. 3), there is valid for their mobility $b = \frac{300 \cdot Q \cdot \log R/r}{2 \cdot \pi \cdot U \cdot I}$ [cgs]

in which Q = total air flow volume; R = inner radius of outer electrode; r = outer radius of inner electrode; U = condenser potential in V; I = distance from input orifice after which the particle reaches the anode.

The individual rings of the anode are thinly coated with vaseline on the inside. At the end of the interval of exposure, the vaseline coating containing the radioactive deposit was wiped off with filter paper moistened in benzene and the activity measured by a β -counter.

Findings

Some preliminary experiments were carried out to test the efficiency of the ionizer. It was necessary to determine the degree of precipitation of the arrangement. Consequently, an electrode filter (Ref. 4) of known efficiency was operated parallel to the cylindrical condenser. After exposure for 24 hours and a condenser potential of 3 kV, an overall efficiency of $64.8 \pm 6.3\%$ for natural activity was found. It was shown that the larger part of the remaining 35% of the activity was precipitated on the walls between the ionizer and at the discharge opening of the ring gap into the condenser. This probably concerns predominantly particles with high mobility which are precipitated in the field between ionizer and intake tube or those

particles coming into contact with the wall due to turbulence. Since the following measurements concern only relative values of activity, the activity precipitated in the ring gap was neglected. The result of determining efficiency indicated that the larger part of the natural activity is found on small medium ions (cf. below).

It would be better in principle to abandon the additional artificial ionization of all particles and to utilize only the natural charge carriers for the count. This reduces the number of particles to one-fourth. At an average activity of 1 to 3 impulses per minute on the individual electrode ring, this was obviously not acceptable. Unfortunately, artificial ionization produces some additional difficulties in the interpretation of the findings because the charge distribution for particles in a corona discharge is not known, in contrast to the natural charge distribution.

Two series of measurements were carried out. The electrode potential with 3 and/or 0.6 kV was selected so that a somewhat larger range of mobility than in the measurements of Wilkening could be detected. Each series consisted of 36 exposures lasting 24 hours. The first series at 3 kV was carried out in March and April 1959 and the second at 0.6 kV in May and June 1959.

The result is shown in Fig. 2. The activity of the individual electrode sections in percent of total activity for each measurement series is represented as function of mobility. The individual anode sections are designated by lower-case Latin letters (the measurement of Wilkening had also been plotted for comparison). In regard to the extent of activity, the two measurement series are comparable only conditionally since the efficiency at 0.6 kV is only about one-half of that at 3 kV. All particles with mobility below 4 cgs-units are no longer precipitated at 0.6 kV. A considerable difficulty in evaluation was represented by the low activities which were in the mean about 10 to 20 impulses per minute for the entire anode. A maximum of 5 impulses per minute was found for 1 ring. Consequently the values show considerable scatter during the individual exposures. The mean errors are plotted in Fig. 2 by thin lines. Wilkening does not indicate any errors. However, they can be estimated from the other data and are of the same order of magnitude as ours. The particle radii for different charge values were calculated with the Stokes-Cunningham formula and plotted in Fig. 2: $b = \frac{n \cdot e (1 - B/pa)}{6 \cdot \pi \cdot \eta \cdot a}$ in which b = mobility; e = elemental charge; p =

air pressure in cm Hg; n = number of elemental charges; a = particle radius; B = constant = 0.000617; η = viscosity of air.

Discussion of Findings

The distribution of activity over the different mobilities initially offers a rather confusing picture. It appears as if, in the range between 1 and 10 cgs, three classes of mobility (h, e and b + e) seem noticeably preferred, even in consideration of the error limits (3-kV series). In the range between 5 and 100 cgs (0.6-kV series), however, a rather uniform distribution was found. Comparison with the measurements of Wilkening

(also plotted) shows preferred regions which concord approximately with our regions j, e and b. When taking into account the error limits, this concordance would not appear to be purely random. In spite of the probably rather different conditions for the origin and composition of aerosols in New Mexico and/or Munich, there consequently results, in the range of the small medium ions, a similar distribution of mobility of natural activity.

However, the objective of our investigations was primarily the range of size of the carriers of natural activity. Prerequisite for this is knowledge of the charge distribution. Wilkening assumes that almost only single charged particles exist in the range of size investigated and cites as proof that no multiple charged particles were manifested in his measurements. This assumption is valid for the natural charge distribution in the atmosphere. However, in the ion atmosphere of a corona discharge, a maximum charge becomes adjusted for particles below a radius of 10^{-4} cm according to the following relation (Ref. 5): $n = 2 \cdot 10^5 \cdot a$.

As will be seen from the scales for the particle radii in Fig. 2, charge 1 (corresponding to a radius of 5×10^{-7} cm) lies exactly in the middle of the range of size investigated. The occurrence of multiple charges is therefore entirely probable. With our arrangements, the particle remained about 5×10^{-3} sec in the corona discharge. This time is sufficient for about 25% of the particles to reach maximum charge. The time between charging and precipitation of the particles lies between 2.5 and 14×10^{-3} sec. It may be assumed that at least a part of the multiple charged particles retains the charge during this time. More detailed indications on the multiplicity and life span of the artificially produced particle charge are only infrequent in the present literature. It would therefore be perhaps best to entirely abandon the artificial ionization of the particles and to restrict the investigation to natural charge carriers. However, this would make it necessary to appreciably enlarge the dimensions of the arrangement. Such investigations are under consideration.

The following table contains the evaluation of our measurements (Fig. 2) by particle size. In consideration of the above statement on charging conditions, the principally occurring particle radii are listed and the electrode sections on which they are found with different charged values are indicated.

Evaluation of Measurements

(a) Teilchenradius in 10^{-7} cm	(b) Vorkommen		
	(c) einfach geladen	(d) zweifach geladen	(e) dreifach geladen
16.5	-	h	•
8.5	•	u	•
4.3	q	m	-
2.3	m	-	-
1.5	k	-	-

a = particle radius in 10^{-7} cm; b = occurrence; c = single charged; d = double charged; e = triple charged.

It will be seen that the viewpoint adopted by Wilkening that the greater part of natural activity is distributed among only two classes of size, i.e. radii of 4.5 and 9×10^{-7} cm, is not confirmed for conditions in Munich. Moreover, it should not be expected that the aerosol of large cities exists in so closely adjacent discrete particle sizes that the radon and thoron metabolons prefer such discrete particle sizes. Further investigations are now to be carried out without artificial ionization and are to extend over a greater range of mobility.

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Figure Appendix

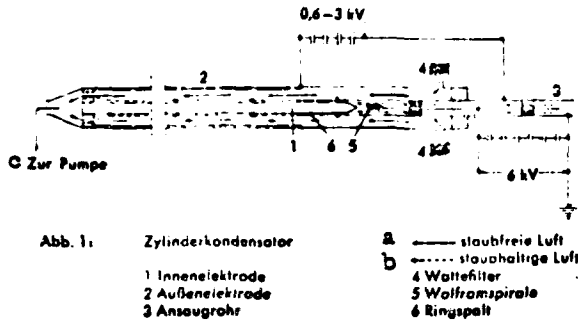


Fig. 1 - Cylindrical condenser

- 1 = inner electrode;
- 2 = outer electrode;
- 3 = intake tube;
- 4 = cotton filter;
- 5 = tungsten coil;
- 6 = ring gap.
- a = dust-free air;
- b = dust-containing air;
- c = to pump.

Abb. 2: Verteilung der Aktivität (in Prozent der Gesamtaktivität jeder Meßreihe) auf die Beweglichkeit.

- a (a-j) I. Serie bei 3 kV
- b ——— (k-u) II. Serie bei 0.6 kV
- c x x x, o o o Meßpunkte von Wilkening
- d r_1, r_2, r_3 Radien für 1-, 2-, 3-fach geladene Teilchen

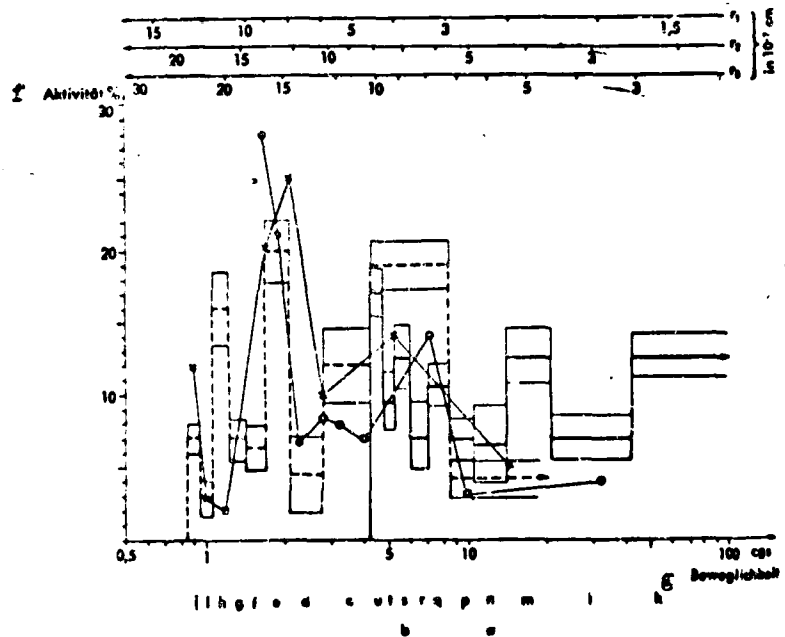


Fig. 2 - Distribution of activity (in percent of total activity of each measurement series) over mobility.

- a = (a-j) 3-kV series; b = (k-u) 0.6-kV series; c = measuring points of Wilkening; d = r_1, r_2, r_3 = radii for single-, double-, triple-charged particles; f = activity; g = mobility.